Supplementary Information: Atomic resolution coherent x-ray imaging with physics-based phase retrieval

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Abstract

Coherent x-ray imaging and scattering from accelerator based sources such as synchrotrons continues to impact biology, medicine, technology, and materials science. Many synchrotrons around the world are currently undergoing major upgrades to increase their available coherent x-ray flux by approximately two orders of magnitude. The improvement of synchrotrons may enable imaging of materials *in operando* at the atomic scale which may revolutionize battery and catalysis technologies. Current algorithms used for phase retrieval in coherent x-ray imaging are based on the projection onto sets method. These traditional iterative phase retrieval methods will become more computationally expensive as they push towards atomic resolution and may struggle to converge. Additionally, these methods do not incorporate physical information that may additionally constrain the solution. In this work, we present a new algorithm which incorporates molecular dynamics into Bragg coherent diffraction imaging (BCDI). This algorithm, which we call PRAMMol (Phase Retrieval with Atomic Modeling and Molecular Dynamics) combines statistical techniques with molecular dynamics to solve the phase retrieval problem. We present several examples where

our algorithm is applied to simulated coherent diffraction from 3D crystals and show convergence to the correct solution at the atomic scale.

Keywords: Phase Retrieval, BCDI, Molecular Dynamics

1 Supplementary Video 1

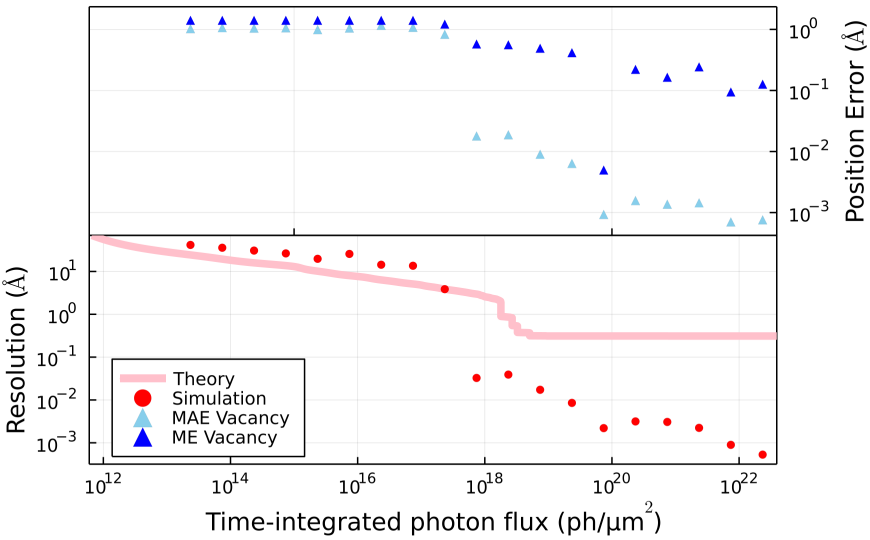
As described in Section 2.2, PRAMMol successfully reconstructs a bulk crystal with no defects. The top left shows the full sample with predicted atom positions (red) compared to the true atom positions (blue), the top center shows the predicted and true central slices for the X-Y, Y-Z, and X-Z planes, and the top right shows a predicted and true central slice of one of the diffraction patterns. Across the bottom (from left to right) shows the predicted and true scaling constant from Equation 2, the predicted and true number of atoms, the error of the predicted diffraction pattern compared to the true diffraction pattern, and the average atom positions error in units of ångströms.

2 Supplementary Video 2

As described in Section 2.2, PRAMMol successfully reconstructs a crystal with a single vacancy. The top left shows the full sample with predicted atom positions (red) compared to the true atom positions (blue), the top center shows the predicted and true central slices for the X-Y, Y-Z, and X-Z planes, and the top right shows a predicted and true central slice of one of the diffraction patterns. Across the bottom (from left to right) shows the predicted and true scaling constant from Equation 2, the predicted and true number of atoms, the error of the predicted diffraction pattern compared to the true diffraction pattern, and the average atom positions error in units of ångströms.

3 Supplementary Video 3

As described in Section 2.2, PRAMMol successfully reconstructs a crystal with a screw dislocation. The top left shows the full sample with predicted atom positions (red) compared to the true atom positions (blue), the top center shows the predicted and true central slices for the X-Y, Y-Z, and X-Z planes, and the top right shows a predicted and true central slice of one of the diffraction patterns. Across the bottom (from left to right) shows the predicted and true scaling constant from Equation 2, the predicted and true number of atoms, the error of the predicted diffraction pattern compared to the true diffraction pattern, and the average atom positions error in units of ångströms.



Supplementary Figure 1 To evaluate the PRAMMol method, 10 trials were run across 20 values of TIPF, for a total of 200 trials. PRAMMol achieves atomic resolution at a TIPF of approximately 10^{18} ph μm^{-2}. This results in resolution and mean average error (MAE) of much less than 1 pm and maximum error (ME) of approximately 1 pm after atomic resolution is achieved.

4 Calculation of Obtainable Resolution vs. Time Integrated Photon Flux

In order to compare our PRAMMol method to traditional iterative phase retrieval algorithms, we followed the methodology of Dietze and Shpyrko to calculate the needed photon flux for resolution scaling with traditional BCDI methods [1]. In Supplementary Section 2.1, we show how PRAMMol compares to the needed photon flux for resolution derived by Dietze and Shpyrko, in section 2.2, we describe our calculation method and in Supplementary Section 2.3, we describe the approximations for the structure factors needed in the diffraction signal calculation.

4.1 Comparison with published work

Supplementary Figure 1 shows that PRAMMol outperforms the predicted iterative phase retrieval resolution scaling theory in two significant ways. First, it is able to achieve atomic resolution with an order of magnitude less TIPF than predicted by traditional phase retrieval theory [1]. Additionally, PRAMMol continues to improve resolution at an accelerated rate as compared to theory.

PRAMMol is able to outperform this theoretical model primarily because a physical model is used in the algorithm. The use of the physical model adds

information about the material system that would ordinarily be difficult to retrieve from a diffraction pattern alone

4.2 Calculation Method

To develop a theory for the photon flux required to achieve a particular resolution, Dietze and Shpyrko assume that the resolution is determined by a cutoff frequency q_c. The diffraction pattern is then sampled across all of reciprocal space that satisfies $\mathsf{q} < q_c$. To determine the total photon flux, Dietze and Shpyrko require that all measurement points have a signal-to-noise ratio greater than or equal to 1. This leads to a relationhip between resolution and time integrated photon flux (TIPF) that is defined in Equation 2 of their paper.

In this work, we use this equation to determine the "theory" line of Fig. 4. However, Dietze and Shpyrko assume that all measurements must be on the Ewald sphere. In our work, we have ignored this experimental detail and have sampled across three disjoint regions of reciprocal space. Additionally, we assume that the polarization factor is always equal to 1 and that the scattering factor does not vary as a function of reciprocal space. While these differences may slightly affect the theoretcial values of TIPF, the results will overall be unchanged, especially the extremely accurate simulation of Fig. 4 where TIPF is greater than 10^{18} photons μm^{-2}.

4.3 Dependence on atomic scattering factors

The diffraction pattern intensity and dependence on the incident photon flux relies upon the atomic scattering factor as described in Section 3.1 of Warren's *X-ray Diffraction* [2]. In Warren, Equation 3.6 describes the dependence of the diffraction signal on the incoming photon flux I_0 and upon the atomic scattering factors F. Here, we assume a small, nanocrystal approximately 6 nm in diameter. Appendix IV in Warren gives the magnitude of the atomic scattering factors as a function of scattering angle (scaled to $sin(\theta)\lambda^{-1}$ where θ is the scattering angle and λ is the wavelength of x-rays). Therefore, we can consider the values of the scattering angle following the assumptions above in Supplementary Section 2.1 of the following parameters: photon energy of 10 keV and $\lambda = 1.24$ Å, detector distance of 1 m, and detector pixel size of 2 mm. Thus, the detector size of the reciprocal space shown in the main text Fig. 3 would be 10 cm on a side.

Furthermore, from Bragg's law, we have $sin(\theta)/\lambda = 1/2d_{\mathrm{hkl}}$ where d_{hkl} is the lattice spacing for the Bragg peak at the hkl reciprocal lattice position. Warren Equation 2.14 gives the value of this lattice spacing for a cubic lattice such as gold as $1/2d^2_{\mathrm{hkl}} = (h^2 + k^2 + l^2)a^{-2}$ where a is the crystal lattice parameter [2]. Therefore, for the reciprocal lattice space cubes considered in the main text and with the parameters described above, we have [h,k,l] values that range from [h,k,l] = [0.4,0.4,0.4] to [1.6,1.6,1.6] and thus $sin(\theta)/\lambda$ ranging from 0.084 $^{-1}$ to 0.340 $^{-1}$. Appendix IV of Warren gives the magnitude of the

atomic scattering factors of gold for $0.1^{-1} = 74.4$ and for $0.4^{-1} = 51.3$. Thus, we conclude that the error in the calculation for the photon flux from one corner of our reciprocal lattice cube to the other to be an approximate maximum factor of $(74.4/51.3)^2 = 2.1$. The square in this calculation is due to the fact that the diffraction pattern intensity calculation has the factor of FF^*, thus the square of the magnitude of the atomic scattering factor. This means that the diffraction patterns used in our simulation and the PRAMMol reconstruction has a slight error of 2 times intensity compared to what we would expect for simulated results, but given the extreme logarithmic scaling of diffraction, we do not expect this to be have a significant effect upon the results of Fig. 4 in the main text and the estimated potential resolution achieved by our PRAMMol technique.

References

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6 Ethics Declarations

The authors declare no competing interests.